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Nano-Structured Exchange-Spring Hard Magnetic Thin Films

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Abstract

In exchange coupled nano-composite magnets, the length scale of the soft phase is limited to twice the domain wall thickness of the hard phase. We investigated the structural and magnetic properties of $SmCo_5$/Co exchange coupled multilayer grown by magnetron sputtering from Sm and Co targets successively at elevated substrate temperature and subsequent in-situ annealing. X-ray diffraction indicates the formation of polycrystalline $SmCo_5$ hard phase. Formation of hard/soft multilayered structure was confirmed through transmission electron microscopy. Magnetic hysteresis measurements showed single phase like behavior, which indicates the soft layer (Co) is well exchange coupled with the neighboring hard phase of $SmCo_5$. Maximum energy product of 15.37 MGOe with $H_C \sim 3.02$ kOe was obtained.

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Keywords: Exchange spring magnet; thinfilm; $(BH)_{max}$.

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1. Introduction

Recently attention is diverted towards nano-composite magnets for further improvement of the magnetic properties of permanent magnetic thin films. In 1991, Kneller [1] proposed the concept of nano-composite magnets, which consists of a hard phase with high coercive field, H_c and a soft phase with high saturation field, J_s, and nano-scale of these phases are coupled via inter-facial exchange interaction. Most important parameters in exchange-spring magnet are relative volume fraction and the geometry of the hard and soft magnetic layers, which determine the nature of magnetization reversal processes and affect the magnet properties such as

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remanent magnetization, M_{rem}, coercive field, H_c and the energy product $(BH)_{max}$. A number of theoretical approaches have been made to address this problem including deriving analytical expression [2], applying micromagnetic modeling [3] as well as first principle calculations [4]. In general, these theoretical approaches derive the critical thickness of the soft layer sandwiched between two hard layers below which the both phases are rigidly exchange coupled and magnetization reversal takes place at same nucleation field, H_N, resulting in a rectangular hysteresis. Although, H_N, depends on the intrinsic magnetic properties of both soft and hard magnetic materials, however theoretically it is predicted that the thickness of the soft layer, t_s, is approximately twice the domain wall width, δ_h, of the hard phase, which is defined by the relation[5]:

$$\delta_h = \pi\sqrt{A_h/K_h} \quad (1)$$

Where, A_h and K_h are the exchange and anisotropy constants of the hard phase, respectively. With parameters for $SmCo_5$, $A_h = 1.2 \times 10^6$ erg/cm and $K_h = 5 \times 10^7$ erg/cm^3, the domain wall width, δ_h can be derived approximately to be 4.86 nm. Above this critical thickness, both hard and soft phases behave as independent components and the magnetization reversal for soft layer takes place at much lower field and the switching is characterized by inhomogeneous reversal.

Though thin film of nano-composite magnets are fabricated by several methods, sputter deposited films are particularly appealing because of the thickness and the interface of the soft and hard phases are easily controllable in nano-meter scale and thus leading to a well defined structure. Experimentally exchange coupling phenomena was observed in sputter deposited bilayer films made of MgO (110)/Cr/Sm_2Co_7(20 nm)/Fe/Cr system with varying thickness of Fe (2.5 nm to 20 nm) [6] and separate switching transition for hard and soft layers was observed with increasing the thickness of the hard and soft layers. Similar results were reported in FePt/Fe exchange-spring system with increasing the thickness of soft layers [7]. Further improvement on both the nucleation field, H_N and hence the $(BH)_{max}$ of the exchange spring magnet was observed in the bilayered system by post annealing the films [8]. This improvement was correlated with the intermixing of Sm-Co with Fe at the interfaces while annealed these films. After post annealing, these films provide a $(BH)_{max}$ of the order of 27.28 MGOe with H_c ~ 6.0 kOe.

In this report, we have grown $SmCo_5$/Co exchange coupled multilayer by successive deposition of Sm and Co layers at elevated substrate temperatures and subsequent annealing at the same temperature. Due to high substrate temperature and long annealing processes, the interface of Sm/Co gets converted into hard phase of $SmCo_5$. With increasing the Co layer thickness, while keeping other parameters constant, the phenomena of exchange coupling was observed. The hard phase formation was confirmed through X-ray diffraction studies. Further confirmation of their microstructure was done by transmission electron microscopy. Magnetic hysteresis measurements showed single phase like behavior, which indicates the soft phase (Co) is well exchange coupled with the neighboring hard phase of $SmCo_5$. The results of $(BH)_{max}$ were compared with variation of both Sm and Co thicknesses.

2. Experimental Procedure

Multilayer films with the structure Cr (10 nm)/ [Sm (t_{Sm} nm)/Co (t_{Co} nm)]25/Cr (20 nm)on Si substrate was fabricated using Ultra High Vacuum (UHV) compatible magnetron sputtering with base pressure better than 5.0×10^{-9} mbar. Sm and Co target were kept at dc power of 10 and 32 watts, respectively. This resulted in growth rate of 3.2 Å /s and 3.8 Å /s for Sm and Co, respectively. During the deposition, the substrate temperature, T_s, was maintained at

450°C and in-situ post deposition annealing was carried out at the same temperature for 1 hr. Several films were grown by varying t_{Sm} from 1.6 to 4.8 nm with t_{Co} at 7.6 nm as well as with varying t_{Co} from 5.7 nm to 11.4 nm with t_{Sm} at 3.2 nm. The film structure was characterized by X-ray diffraction and cross-sectional Transmission Electron Microscopy (TEM). The magnetic properties at room temperature were measured using Vibrating Sample Magnetometer (VSM).

3. Result and Discussion

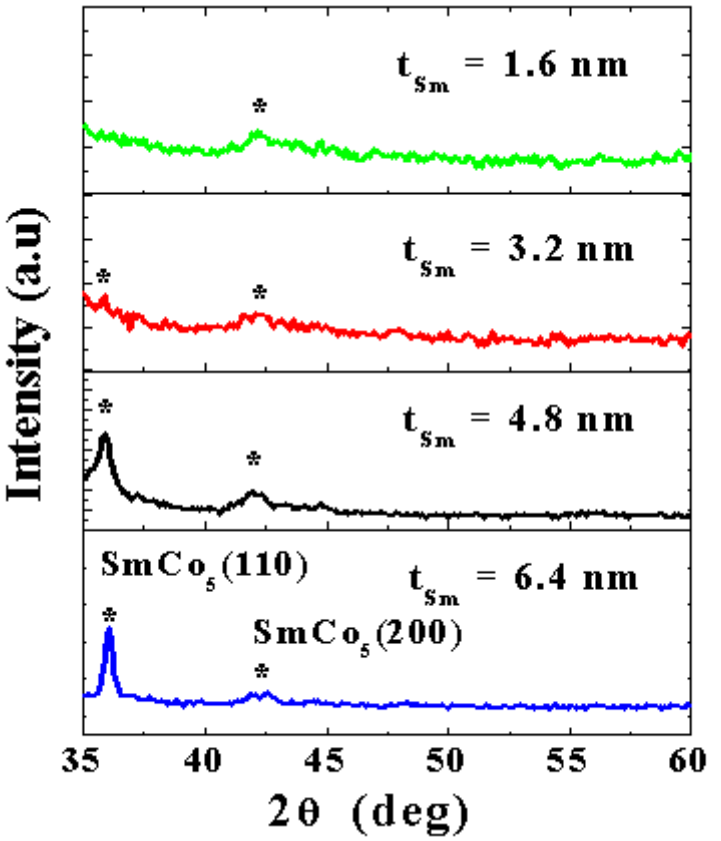


Fig. 1 (a) X-ray diffraction patterns of $SmCo_5$/Co multilayer films as a function of t_{Sm} at constant t_{Co} .

The X-ray diffraction patterns with varying Sm thicknesses are shown in Fig. 1a. For $t_{Sm} < 3.2$ nm, films are amorphous under x-ray diffraction. With increasing thickness, the peaks corresponding to $SmCo_5$(110) and (200) appear. This indicates that the crystalline growth of $SmCo_5$ phase with increase in the Sm thickness.

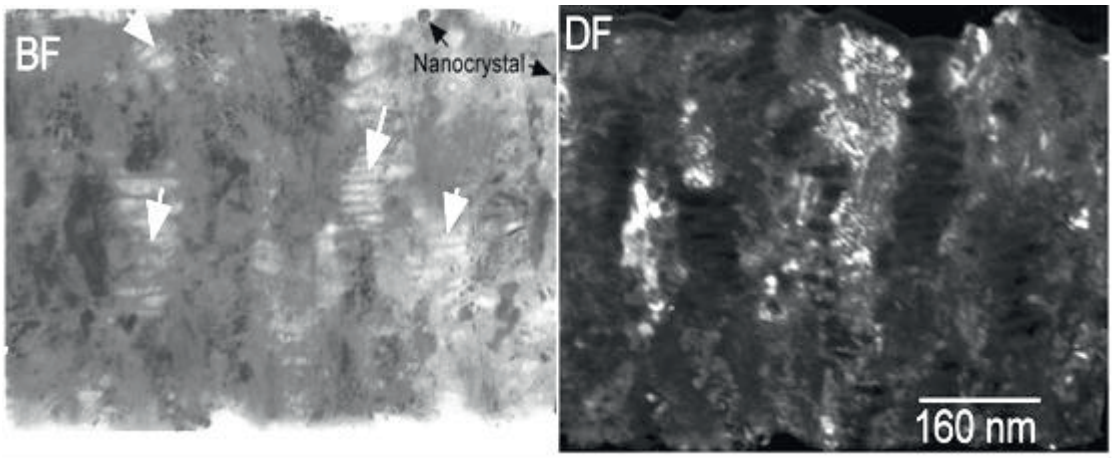


Fig. 1.b 2 TEM images for films with structure Cr (10 nm)/[(Sm (3.2 nm)/Co(5.7 nm)]$_{25}$/Cr(20 nm)/Si.

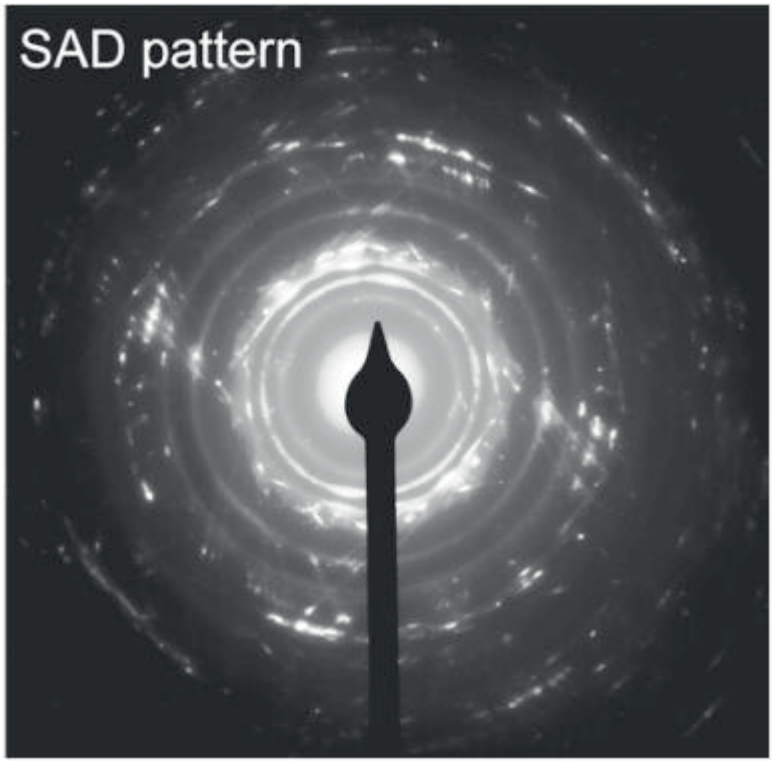


Fig. 1c Selected area electron diffraction pattern of the film with above mentioned structure.

Further investigation was performed by Transmission Electron Micrograph (TEM) to study the structure of the film. Fig. 1(b) shows the bright field (BF), as well as dark field (DF) cross sectional TEM image of a film of structure Cr(10 nm)/[(Sm (3.2 nm)/Co(5.7 nm)]25/Cr(20 nm)/Si prepared at a substrate temperature of 450^{o}C and annealed at the same temperature for 1 hr. The bright field cross- sectional image shows the formation of scattered layered structure with well defined boundaries between the hard and soft phases, indicated by white arrows (see Fig:1(b) (left)). From this multilayered structure the total thickness of hard and soft layer together was found to be around 9 nm, which was similar to the predicted thickness calculated according to the growth rate of Sm and Co, respectively. Along with these multilayered structures, formation of nanocrystalline structure was also observed as shown by dark arrows. This might be due to the high reactivity of Sm with Co at higher deposition temperatures. Well defined multilayered structure can be formed by increasing the Co layer thickness. The combined selected area electron diffraction (SAED) pattern indicates the polycrystalline growth of $SmCo_5$ phase as shown in Fig 1(c).

Magnetic hysteresis loops are measured by Vibrating Sample Magnetometer to show the effect of layer thicknesses on magnetic properties. The film layer structure was Cr(10 nm)/[(Sm (t_{Sm} nm)/Co(t_{Co} nm)]$_{25}$/Cr(20 nm)/Si. Fig. 2 shows the M(H) loop for samples with varying t_{Co} while t_{Sm} was kept constant and Fig. 3 shows the same for samples with varying t_{Sm} at constant t_{Co} value. It is to be mentioned that all the M(H) hysteresis loops measurements were carried out at room temperature and with field parallel to the film plane. The behavior of M(H) loops indicates the soft and hard layers coupled rigidly.

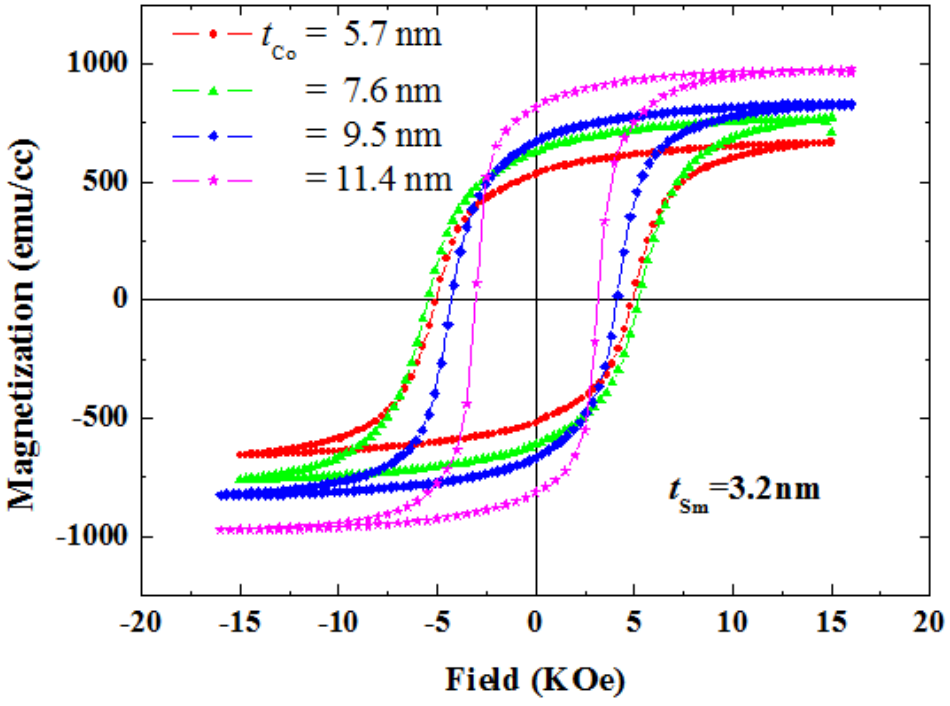


Fig 2. M(H) hysteresis loops with varying the Co layer thickness keeping t_{Sm}= 3.2 nm

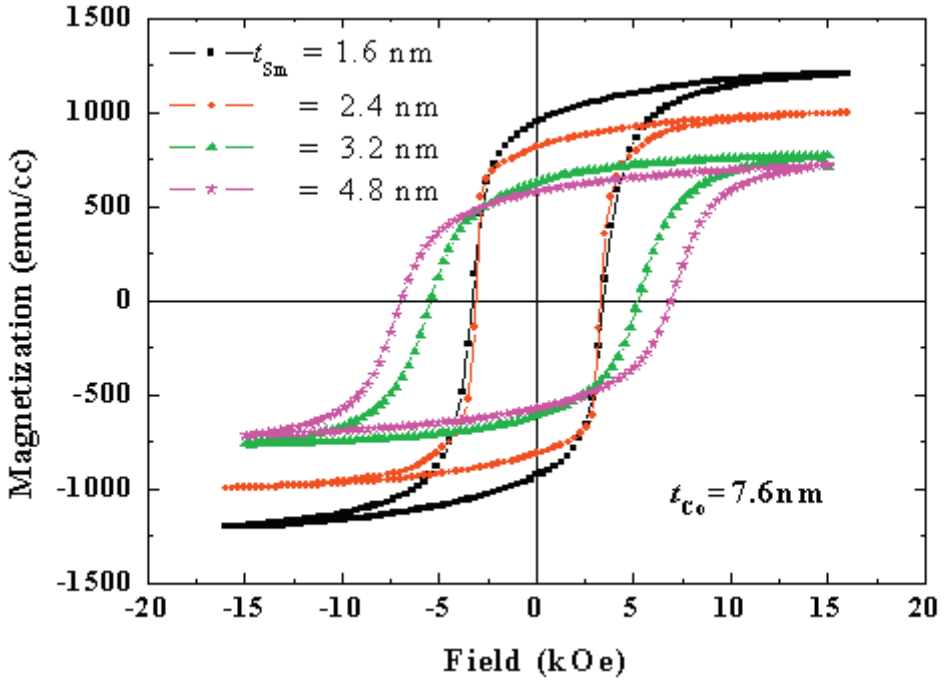


Fig. 3 M(H) hysteresis loops with varying the Sm layer thickness keeping t_{Co} = 7.6 nm.

The magnetic parameters, such as total saturation magnetic moment, M_{res}, squareness ratio S, coercivity, H_c and the energy product, $(BH)_{max}$ obtained from these figures are tabulated in Table 1. As given in this table, S is almost independent of layered structure for the samples studied here.The estimated values of $(S > 0.8)$ are similar to those reported for SmCo/Co systems [10,11]. It is interesting to see that for constant t_{Sm}, M_{res} value increases with increase in t_{Co}, whereas it decrease with an increase in t_{Sm} for constant t_{Co}. The behavior of M_{res} depends strictly on the volume contribution of individual soft and hard layers and this can be correlated as[5]:

$$M_{res} = \frac{(M_h t_h + M_s t_s)}{(t_h + t_s)} \qquad (2)$$

With, $t_h + t_s = t_{Sm} + t_{Co}$, the above equation leads to

$$t_s = \frac{t_{Sm} + t_{Co}}{\left(1 + \frac{M_s - M_{res}}{M_{res} - M_h}\right)} \qquad for\ t_{Sm} < t_{Co} \qquad (3)$$

And,

$$t_h = (t_{Sm} + t_{Co}) - t_s \qquad (4)$$

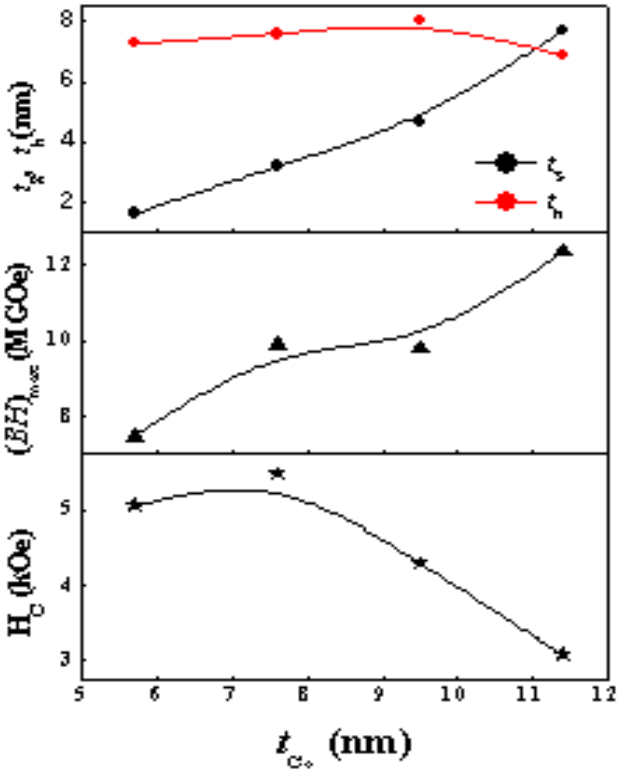
The magnetic moment, M_s, of pure Co film was measured to be 1407.08 emu/cc. The value is close to 1400 emu/cc as reported by other authors [9]. This resultant saturation magnetic moment, M_{res},is lower, since $SmCo_5$ phase has lower magnetic moment, M_h, of the

order of 500 emu/cc[3]. With following parameters, M_s = 1400 emu/cc, M_h = 500 emu/cc and experimentally measured M_{res}, one can determine the thickness of hard and soft layers using equations (3) and (4).

Table 1. Magnetic parameters calculated from MH loops with varying Sm and Co thicknesses.

t_{Sm} (nm)	t_{Co} (nm)	H_C (kOe)	M_{rem} (emu/cc)	M_{Sat} (emu/cc)	S (M_{rem}/M_{sat})	$(BH)_{max}$ (MGOe)
	5.7	5.06	531.90	663.39	0.80	7.46
3.2	7.6	5.47	629.30	766.97	0.82	9.89
	9.5	4.28	672.60	829.23	0.81	9.80
	11.4	3.07	814.40	975.03	0.84	12.38
1.6		3.02	947.5	1203.12	0.79	15.37
2.4	7.6	3.13	813.70	995.01	0.82	14.44
3.2		5.47	629.30	766.97	0.82	9.89
4.8		6.98	578.60	716.65	0.81	9.68

(a)



(b)

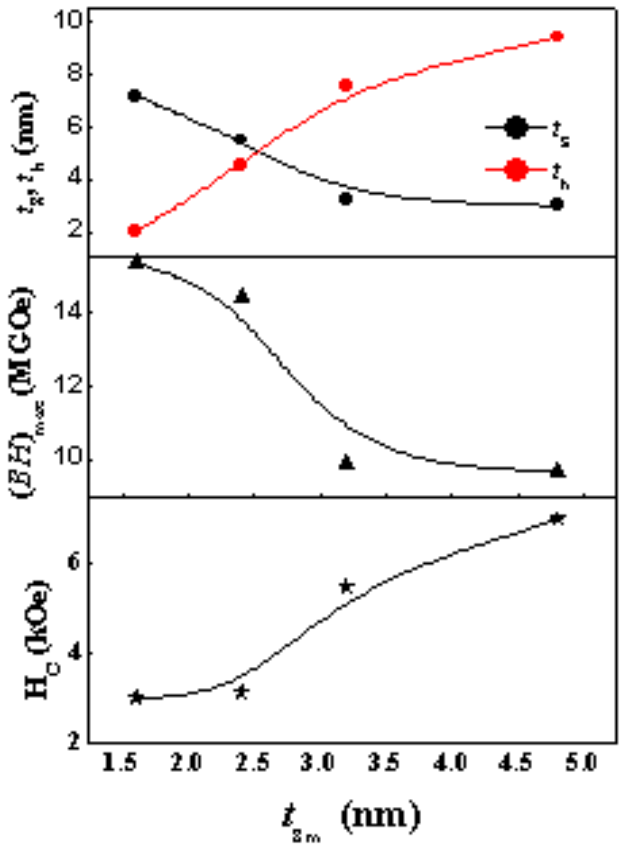


Fig 4 Variation of hard(t_h) and soft(t_s) layer thicknesses, $(BH)_{max}$ and H_C with varying t_{Co} at

v

In addition, it implies that the hard layers thickness can be controlled judiciously by varying the thickness of Sm layers. Therefore, this is an effective method for growth of exchange coupled magnetic thin films with required magnetic properties. It will be interesting to study the X-ray reflectivity to provide a quantitative agreement with these estimated values of t_s and t_h.

$(BH)_{max}$ and H_C as a function of t_{Co} and t_{Sm} are shown in Fig 4(a) and 4(b), respectively. As shown in Fig 3(a), decrease in H_c from 5.4 kOe to 3.0 kOe with an increase in t_{Co} is due to the reduction of exchange coupling with increasing of t_s. The opposite cases were observed with increasing t_{Sm} as shown in Fig 3 (b), where the hard phase is dominating and exchange coupling was enhanced. In the case of $(BH)_{max}$, there is an increase from 7.46 MGOe to 12.38 MGOe for varying Co thickness from 5.7 nm to 11.4 nm, respectively due to increase in magnetic saturation value. However, with increasing Sm thickness, $(BH)_{max}$ value was found to decrease. It is interesting to see that highest $(BH)_{max}$ of 15.37 MGOe was obtained for t_{Sm} = 1.6 nm and t_{Sm} = 7.6 nm with 3.02 kOe which is reasonably high for any permanent magnetic thin films.

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